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TECHNICAL DIVISION
SAVANNAH RIVER LABORATORY

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CC: J. W. Croach-A. A. Johnson, Wilm.
W. B. DeLong
W. P. Bebbington, SRP
T. C. Evans
E. B. Sheldon
R. F. Rogers-J. C. Eargle
J. P. Maloney-E. S. Bridges
C. H. Ice-L. H. Meyer, SRL
H. J. Groh
S. Mirshak
R. T. Huntoon
P. K. Smith
D. T. Rankin
C. L. Selby
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M E M O R A N D U M

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TO: J. M. BOSWELL

FROM: R. GREGG RG.

FABRICATION AND EXTRUSION OF NpO_2-Al BILLETS

INTRODUCTION AND SUMMARY

Tubular ^{237}Np target elements are fabricated by coextrusion of Al billets with cores of compacted Al-NpO_2 powders. This memorandum documents studies of the effect of lubricant composition on core compaction and reviews the effect of Np concentration and Al-NpO_2 interaction on extrusion behavior. The information presented provides bases for limits of lubricant composition and Np concentration for compaction and extrusion of NpO_2-Al cores.

Studies at SRL demonstrated that concentrations of aluminum stearate $>$15 wt % in dodecanol do not appreciably improve lubricity during compaction and ejection of core compacts, as illustrated in Figure 1. Hence, the concentration of aluminum stearate in dodecanol should be limited to $\approx$15 weight % to minimize the time required for billet outgassing and the amount of solid residue in the outgassed billet.

Cracking can occur during drawing of NpO_2-Al tubes in core areas containing approximately 35-40 volume % of hard phases. This level of hard phase concentration can be obtained in target cores containing only 12 volume % NpO_2 if complete reduction of NpO_2 by the Al matrix occurs during billet outgassing. A correlation is shown between the extent of Al-NpO_2 interaction and the deformation pattern during powder compaction. To eliminate core cracking experienced with Mark 52, as a result of hard phase formation, the Mark 53 target was designed for billet core compacts of 80% theoretical density (compared to 90% for Mark 52) and a NpO_2 concentration of $\approx$6 volume % (12.3% for Mark 52).

DISCUSSION

Core Compaction - Lubrication

Billet cores of Mark 52, 53, and 61 Np targets are fabricated by compaction of blended NpO_2 and Al powder blends to final core dimensions. A suspension of finely divided, solid aluminum stearate in liquid dodecanol is used for lubricating the die and punch prior to compaction.(1) The lubricant retained in the compacts adds to the gas load to be removed during billet outgassing and can increase outgassing time and subsequently, formation of hard spots from Al-NpO_2 interaction as discussed below. A solid residue, primarily from the stearate, remains in the billet after outgassing.(2) The amount of residue is small and no problems are known to result from its presence; however, significantly greater stearate concentrations could result in interference with core-clad bonding during tube coextrusion.

Tests were made with the Mark 53 compaction tooling to evaluate the effect of stearate concentration on reduction of compaction and ejection stresses. Results of the tests are illustrated in Figure 1. Compaction to two final powder densities was evaluated; 88 and 90% theoretical density (3.415- and 3.00-inch-long compacts, respectively). In production, NpO_2-Al compacts are pressed to 80% density. ThO_2 was used as stand-in for NpO_2 and at a volume concentration corresponding to the maximum NpO_2 concentration used in previous Np target tube production (12.3 volume % in Mark 52). Al compacts were also made. Compaction stresses are lower without oxide but galling is normally more severe. Lubricant was prepared using Witco aluminum stearate No. 18 and reagent grade dodecanol. The suspension was stirred and maintained at 75-80°F. A thin film of lubricant was applied to tooling by wiping with a lubricant soaked sponge.

The results, Figure 1, show a decrease in both compaction and ejection stresses* up to 15 weight % stearate. Further additions of stearate could provide increased assurance of desired lubricity in core fabrication but will add undesirably to the amount of volatile material to be removed during billet outgassing.

Extrusion Behavior

^{237}Np target tubes are fabricated by the process used for all coextruded fuel and targets. Billets are outgassed at ≈525°C, lubricated, reheated to ≈425°C for hot extrusion and subsequently cold-drawn and straightened to final dimensions.

Difficulties were experienced with the original NpO_2-Al core extrusions (Mark 52) due to the formation of hard, brittle areas in the core from the reaction(3)

$$Al + NpO_2 \rightarrow NpAl_4 + Al_2O_3$$

* Calculated by dividing press ram force by compact cross-sectional area and die-compact solid contact area, respectively.

Subsequent studies[4] showed that the extent of the reaction increased with time at outgassing temperature and the degree of deformation imparted to the Al powder during compaction.

The latter effect results from the diffusion barrier to the reaction provided by the Al_2O_3 film on each Al powder particle. The Al-NpO_2 interaction can be prevented completely in loosely compacted powders. However, in large high density compacts in which the Al is subjected to significant macroscopic shear deformation, intimate contact is provided between the Al and NpO_2 and the reaction proceeds rapidly.

The location of hard, reacted areas in the extruded core correlate with the deformation pattern in the compact. A schematic of relative densities produced in a compact is shown in Figure 2. Reaction occurs most easily in the high density (deformation) area around the ends of the compact, as shown in the extruded core of Figure 3. This core was outgassed for ≈25 hours at 525°C and only partial reaction occurred. The separation of reacted areas occurred during extrusion (extrusion ratio ≈18:1) and indicates the degree of embrittlement.

At longer outgassing times, the reaction extends to lower density areas, following the deformation pattern of the compact. A cross section through the middle of a compact in an extruded Mark 52 tube is shown in Figure 4. This core was outgassed, prior to extrusion, for ≈95 hours at 525°C. The area in the extruded core corresponds to a cross section at mid-height through the density profile in Figure 2. The reaction which occurred in the compact is confined to the higher density area of the cross section.

Partial, localized NpO_2-Al interaction results in core thickening and nonuniform distribution of ^{237}Np in the tube core. Complete reaction significantly increases the volume of the core occupied by hard, brittle phases, Figure 5, and reduces core ductility. Cracking of the core has been observed during cold-drawing (at ambient temperatures) of Mark 52 tubes in which nearly complete reaction has occurred, Figure 6. Complete reaction of this 12.3 volume % NpO_2 core corresponds to ~40 volume % reaction products.

At extrusion temperatures (≈425°C), accommodation of hard phases is increased by increased ductility. However, significant increases beyond ≈40 volume % would be expected to result in severe core cracking and cladding tearing during extrusion.

The Mark 53 target was designed to reduce and accommodate the core hardening from Al-NpO_2 interaction.[3] Compact density was reduced from ≈90% (Mark 52) to 80% theoretical and the NpO_2 concentration reduced from 12.3 to 6 volume % at a Np loading of 150 g/ft.

RG:ehj

REFERENCES

1. DPSTS-235-F-NC, Technical Standards, Np Billet Fabrication.
2. DP-69-1-3, SRL Monthly Report.
3. DPST-68-605, Design of a Coextruded Neptunium Target Compatible with USH, G. L. Tuer to S. Mirshak, December 12, 1968.
4. DP-69-1-1, SRL Monthly Report.

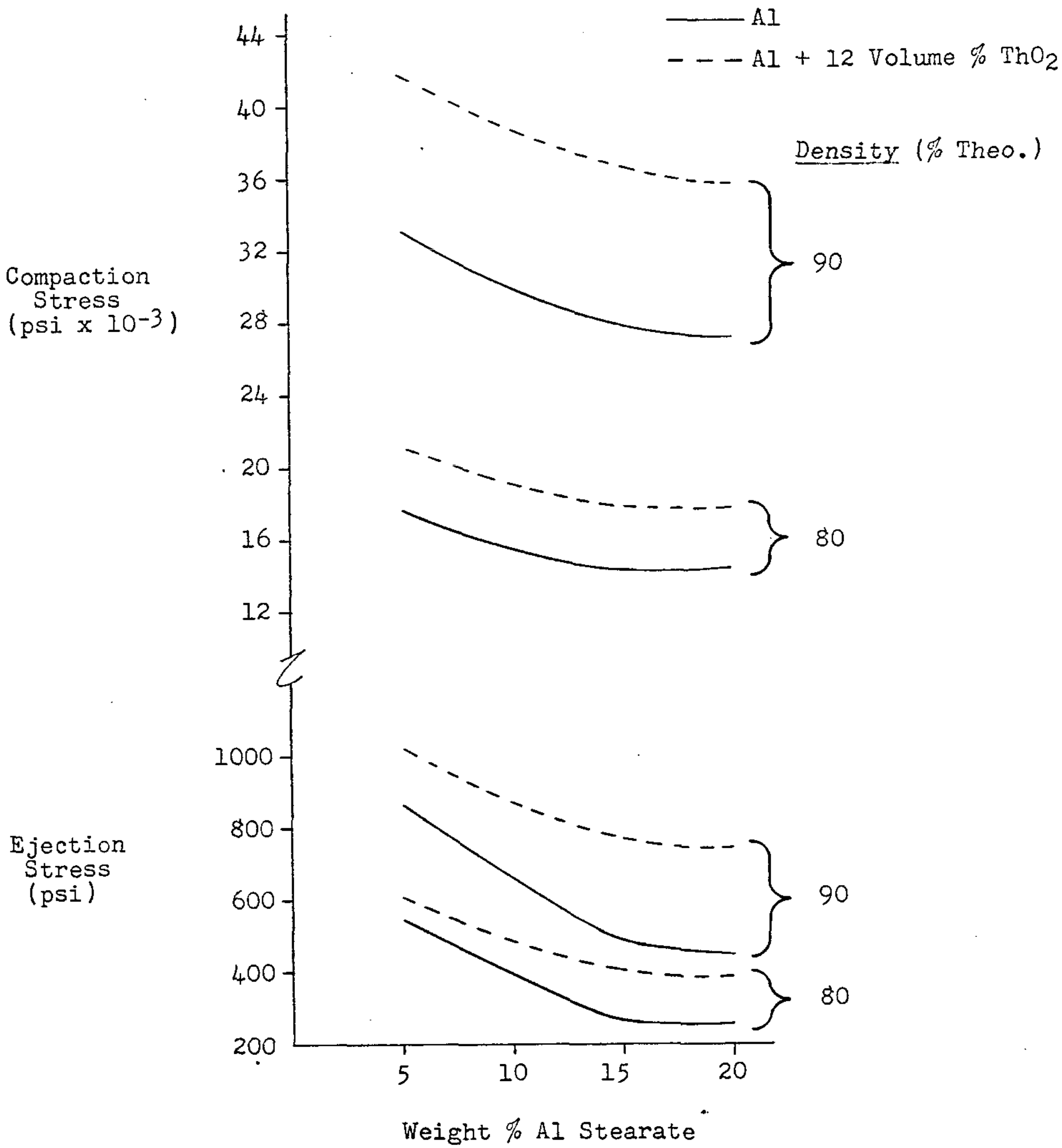

FIGURE 1. EFFECT OF STEARATE CONCENTRATION ON LUBRICITY OF STEARATE-DODECANOL MIXTURES.

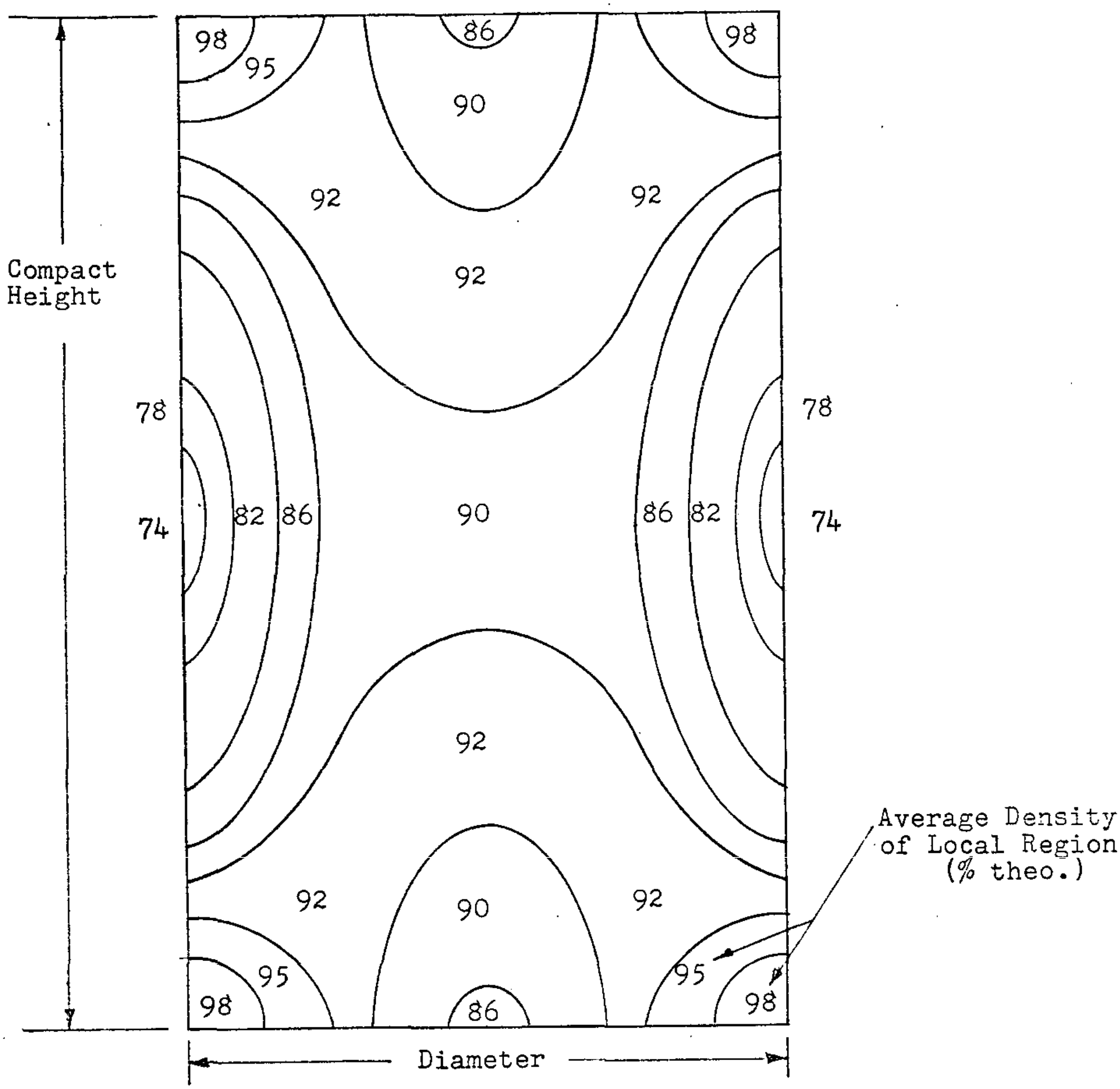

FIGURE 2. SCHEMATIC OF DENSITY PROFILE ON LONGITUDINAL SECTION OF COMPACT PRESSED FROM BOTH ENDS TO AN AVERAGE DENSITY OF ≈90% THEORETICAL.

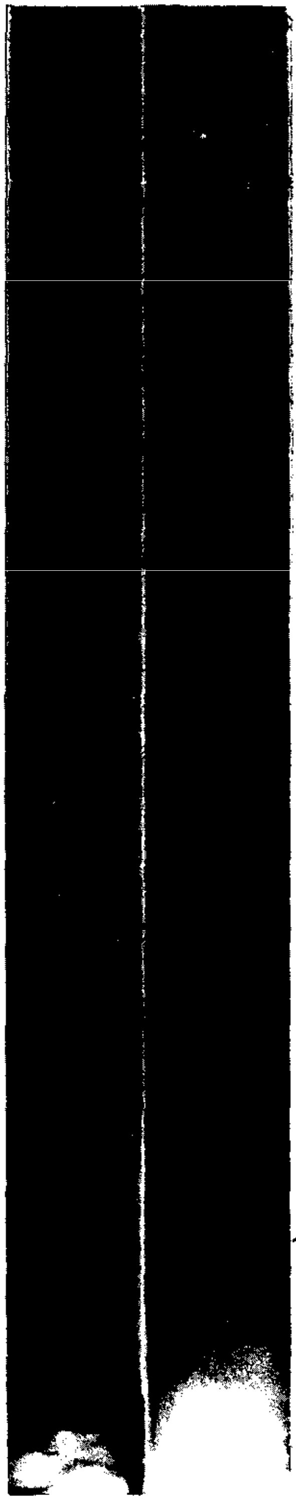

FIGURE 3. ENDS OF TWO EXTRUDED CORE COMPACTS IN MARK 52 TARGET WITH INCOMPLETE Al-NpO_2 REACTION. (dark regions) (Positive print of radiograph)

FIGURE 4. SECTION OF MARK 52 TARGET FROM MIDDLE OF EXTRUDED CORE. Note Al-NpO_2 reaction is confined to center of core compact.

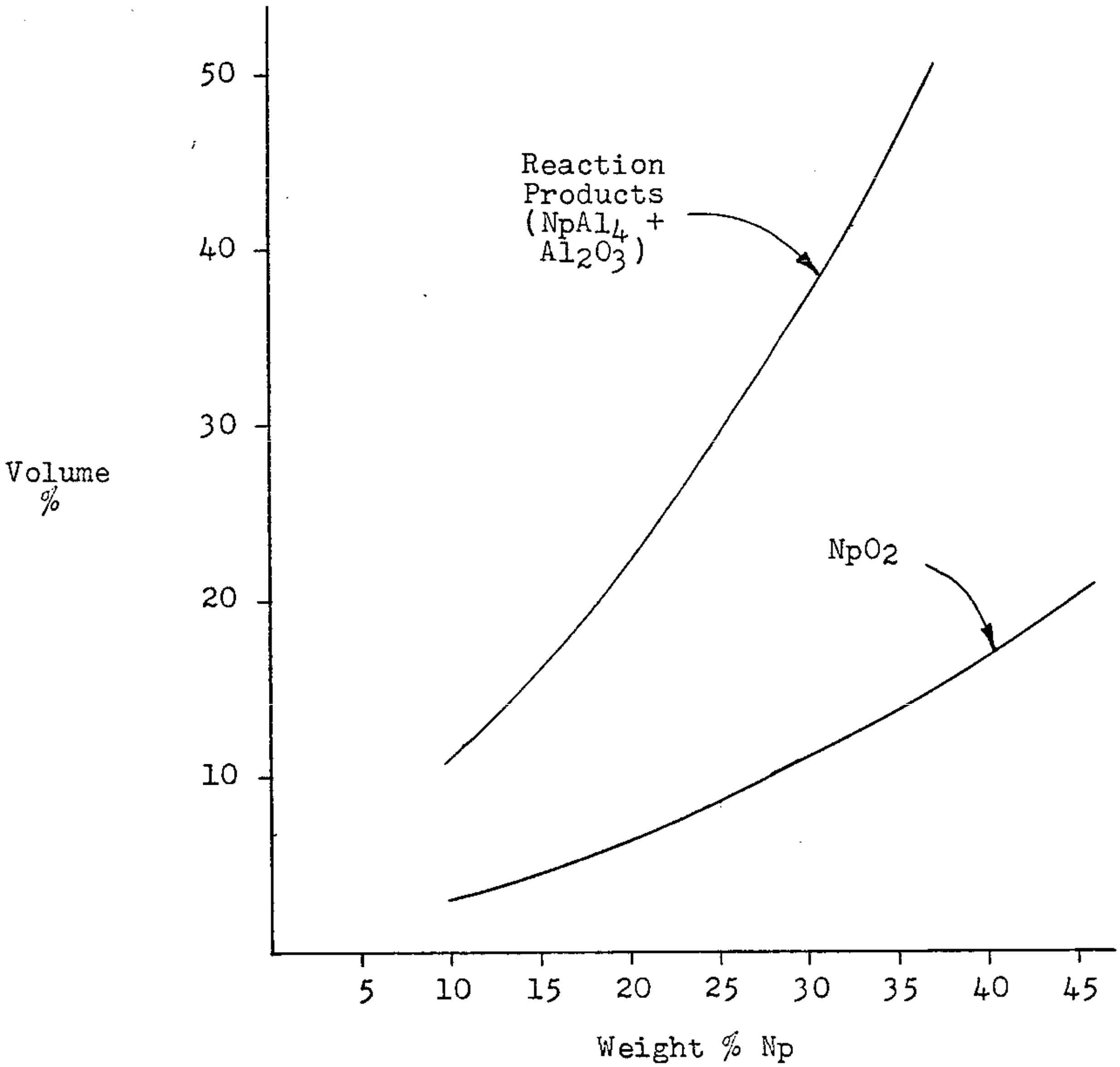

FIGURE 5. HARD PHASE CONTENT FOR MIXTURES OF Al AND NpO_2 POWDERS.

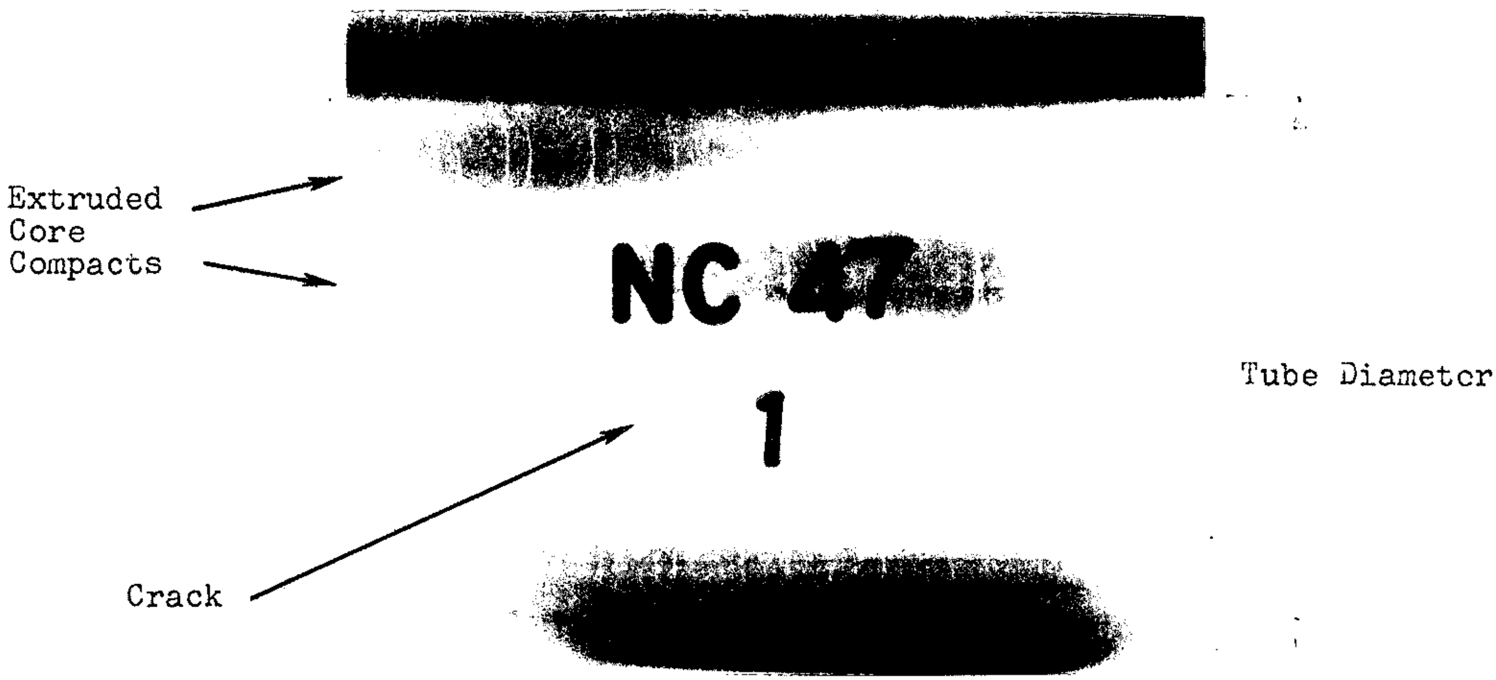

FIGURE 6. CORE CRACKS IN MARK 52 TARGET AT CORE END EMBRITTLED BY Al-NpO_2 REACTION (positive print of radiograph).